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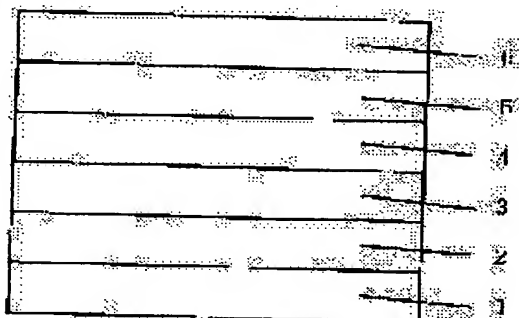
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(54) OXIDE ELECTRODE THIN FILM

(57)Abstract:

PROBLEM TO BE SOLVED: To improve residual polarization characteristics in a ferroelectric capacitor type memory using PZT or the like by a method which is comprised of epitaxially growing a ferroelectrics having a perovskite-type structure and an oxide lower electrode in the (100) or (001) direction on a (100) silicon substrate although it has been hitherto impossible to allow the perovskite-type oxide to orient in (100) or (001) direction on a (100) silicon substrate or on a (100) fluorite structure buffer layer.

SOLUTION: A ferroelectric capacitor of (100) or (001) orientation is realized by epitaxially growing an MO layer 3 (M=Ca, Sr, Ba) comprising an MO layer being part of the perovskite structure and having NaCl structure in the (100) direction on a (100) silicon substrate 1 or on a (100) fluorite structure buffer layer 2 and then successively depositing a perovskite oxide electrode thin film 4 such as SrRuO_3 , a ferroelectric layer 5 and an oxide electrode thin film 6.



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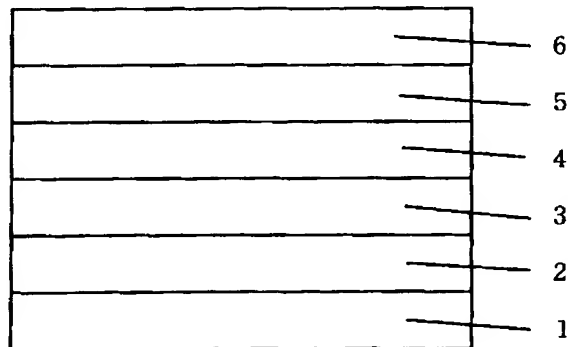
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(54) 【発明の名称】 酸化物電極薄膜

(57) 【要約】

【課題】 P Z T などを用いた強誘電体キャパシタ型メモリにおいて、残留分極特性を向上させるためには、(100) シリコン基板上にペロブスカイト構造を有する強誘電体および酸化物下電極を (100) あるいは (001) 方向にエピタキシャル成長させる必要があるが、従来、(100) シリコン基板上あるいは (100) フルオライト構造バッファ層上に、ペロブスカイト酸化物を (100) あるいは (001) 配向させることは不可能であった。

【解決手段】 (100) シリコン基板1上、あるいは (100) フルオライト構造バッファ層2上に、ペロブスカイト構造の一部であるMO層からなるNaCl構造のMO層3 (M=Ca、Sr、Ba) を (100) 方向でエピタキシャル成長させ、その後、SrRuO₃などのペロブスカイト酸化物電極薄膜4、強誘電体層5、酸化物電極薄膜6を堆積することにより、(100) あるいは (001) 配向の強誘電体キャパシタを実現する。



【特許請求の範囲】

【請求項1】 (100)シリコン基板と、前記シリコン基板上に少なくともNaCl構造の(100)配向した金属酸化物MO (M=Ca、Sr、Ba)を含むバッファ層と、前記バッファ層上にペロブスカイト構造の擬立方晶で(100)配向に相当する金属酸化物層ABO₃とを有することを特徴とする酸化物電極薄膜。

【請求項2】 前記バッファ層は、フルオライト構造の(100)配向した金属酸化物からなる第1バッファ層と、前記第1バッファ層上にNaCl構造の(100)配向した金属酸化物MO (M=Ca、Sr、Ba)からなる第2バッファ層とからなることを特徴とする請求項1記載の酸化物電極薄膜。

【請求項3】 前記第1バッファ層は、(100)配向したイットリア安定化ジルコニアZr_{1-x}Y_xO_{2-0.5x} (0<x<0.45)と(100)配向した酸化セリウムCeO₂からなることを特徴とする請求項2記載の酸化物電極薄膜。

【請求項4】 前記金属酸化物層ABO₃は、MRuO₃ (M=Ca、Sr、Ba)からなることを特徴とする請求項1記載の酸化物電極薄膜。

【請求項5】 前記金属酸化物層ABO₃は、La_{1-x}Sr_xVO₃ (0.23<x<1)からなることを特徴とする請求項1記載の酸化物電極薄膜。

【請求項6】 前記金属酸化物層ABO₃は、La_{1-x}Sr_xMnO₃ (0.2<x<0.4)からなることを特徴とする請求項1記載の酸化物電極薄膜。

【請求項7】 前記金属酸化物層ABO₃は、La_{1-x}Sr_xCoO₃ (0.4<x<0.6)からなることを特徴とする請求項1記載の酸化物電極薄膜。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】本発明は、酸化物電極薄膜に関し、特に、この酸化物電極薄膜を用いた強誘電体メモリなどの酸化物デバイスに関する。

【0002】

【従来の技術】近年、強誘電体を用いた不揮発性メモリの開発が急速に進展している。強誘電体としては、PbZr_{1-x}Ti_xO₃ (PZT)、Ba_{1-x}Sr_xTiO₃ (BST)、SrBi₂Ta₂O₉ (SBT)等の材料が注目されているが、中でも菱面体-正方晶相境界 (MPB) 付近の組成のPZT材料が、その残留分極、抗電界特性に優れており、最も実用化の進展している材料である。

【0003】強誘電体メモリは、強誘電体をキャパシタとして用い1T/1C構造などを形成する強誘電体キャパシタ型と、強誘電体をSiO₂の代わりにゲート絶縁膜として用いるMFSFET型とに分類される。1T/1C型の強誘電体キャパシタ構造においては、シリコン(Si)などの基板上に電極層を形成し、その上に強誘電体層を形成することになるので、電極材料の選択が強

誘電体の特性に大きな影響を与える。強誘電体キャパシタの電極材料として要求される特性は、

- (1) 電気抵抗が十分低い。
 - (2) 強誘電体材料との格子定数のミスマッチが小さい。
 - (3) 耐熱性が高い。
 - (4) 反応性が低い。
 - (5) 拡散バリア性が高い。
 - (6) 基板、強誘電体との密着性が良い。
- などである。

【0004】従来、PZT系強誘電体の電極材料として用いられてきたのはPtである。Ptは最密充填構造である面心立方格子(FCC)構造をとるため自己配向性が強く、SiO₂のようなアモルファス上に成膜すると(111)に強く配向し、その上の強誘電体膜も配向性が良い。しかし、配向性が強い柱状結晶が成長し、粒界に沿ってPbなどが下地に拡散しやすくなるといった問題があった。またPtとSiO₂との密着性にも問題があった。

【0005】ここでPtとSiO₂との密着性の改善のためにTi、さらにPbなどの拡散バリア層としてTiNなどを用いても、複雑な電極構造になる上、Tiの酸化、TiのPt中への拡散、それに伴うPZTの結晶性の低下が起こり、P-Eヒステリシス特性、リーク電流特性、ファティーグ特性などの電気特性が劣化する。

【0006】このようにPt電極には多くの問題があるため、RuO_xやIrO₂をはじめ、導電性酸化物電極材料が研究されている。中でも、ペロブスカイト構造を有するSrRuO₃は、PZTと同じ結晶構造を有しているので、界面での接合性に優れ、PZTのエピタキシャル成長を実現しやすく、Pbの拡散バリア層としての特性にも優れている。従って、SrRuO₃を電極として用いた強誘電体キャパシタの研究が盛んに行われている。

【0007】

【発明が解決しようとする課題】しかし、SrRuO₃電極を酸化物電極薄膜、PZTを強誘電体として強誘電体キャパシタを構築する場合、以下のような問題があった。

【0008】PZTは、Zr:Ti=0.48:0.52のMPBよりTi過剰側の例えばZr:Ti=0.3:0.7、0.2:0.8の組成が、残留分極P_rの増大、抗電界E_cの低減の点から重要である。またこの組成領域でのPZTは正方晶を示し、その分極方向はc軸に平行である。従って、上部電極/強誘電体/下部電極/基板の構造を有する強誘電体キャパシタでは、強誘電体層PZTの(001)配向膜を得るため、下部電極であるSrRuO₃電極自体を(100)方向(擬立方晶)に配向成長させる必要がある。

【0009】ところが、Si上にペロブスカイト型酸化

物である SrRuO_3 電極を直接エピタキシャル成長させることは、困難である。そこで、 Si 上に何らかのバッファ層をエピタキシャル成長させ、その上に SrRuO_3 電極をエピタキシャル成長させる必要がある。

【0010】ここで、 Si 上にペロブスカイトと類似の構造を有する $\text{YBa}_2\text{Cu}_3\text{O}_x$ をエピタキシャル成長させるためのバッファ層としては、イットリア安定化ジルコニア $\text{Zr}_{1-x}\text{Y}_x\text{O}_{2-0.5x}$ (YSZ)、 CeO_2 、 Y_2O_3 などフルオライト構造を有する酸化物が用いられてきた。また、 $\text{Y}_2\text{O}_3/\text{YSZ}$ ((1) Appl. Phys. Lett., 61 1240 (1992))、 CeO_2/YSZ ((2) Appl. Phys. Lett., 64 1573 (1994)) の二重バッファ層も $\text{YBa}_2\text{Cu}_3\text{O}_x$ をエピタキシャル成長させることが確かめられている。この場合 $\text{YBa}_2\text{Cu}_3\text{O}_x$ は容易に(001)配向する。

【0011】ところが、単純ペロブスカイト構造である SrRuO_3 (擬立方晶において $a=0.393\text{nm}$ 、 $2^{1/2}a=0.556\text{nm}$) の場合は、フルオライト型構造の YSZ ($a=0.514\text{nm}$) あるいは CeO_2 ($a=0.541\text{nm}$) の(100)面上には、格子の不整合は CeO_2 の場合で3%程度と小さいに関わらず、(100)方向にエピタキシャル成長しない。これは、(100)配向のフルオライト構造においては、最表面が Ce イオンのみまたは酸素イオンのみの電荷が中和されない面であるのに対し、(100)配向のペロブスカイト型構造では、各 AO 、 BO_2 層ともに陽イオン、陰イオンからなる電荷の中和された面であり、下地の最表面と堆積層の第一層との静電相互作用によって、反発力が生ずるためである。

【0012】例えば、(100) YSZ 基板には(110)配向 (擬立方晶) することが知られている ((3) Appl. Phys. Lett., 67 1387-1389 (1995))。これは、(100)配向のフルオライト構造の最表面に(110)配向のペロブスカイト構造が積層する場合、(110)面方向に ABO 面、 O_2 面が積層する構造であり、各面の電荷が中和しておらず静電相互作用の観点で安定であるために、(100)配向のフルオライト構造の上に容易に配向するものと考えられる。

【0013】さらに、(100) $\text{SrRuO}_3/(100)\text{CeO}_2/(100)\text{Si}$ の酸化物積層構造および製造方法が提案されている ((4)特開平10-182292)。しかしながら実際には(100) $\text{CeO}_2/(100)\text{Si}$ のエピタキシャル成長は、600℃以上の高温かつ 10^{-8}Torr 以下の高真空雰囲気下でしか可能でなく、しかも単一配向膜にはならず、(110) $\text{CeO}_2/(100)\text{Si}$ の領域と共存する ((5)Jpn. J. Appl. Phys., 29, L1199 (1990))。また(100) $\text{SrRuO}_3/(100)\text{CeO}_2/(100)\text{YSZ}/(100)\text{Si}$ の SrRuO_3 にも(110)方向と(100)方向とが共存している。

【0014】以上のように、 Sr と Ru が同時供給されて成長する限りは、 Si 基板上あるいはフルオライト構造のバッファ層上に、(100)配向のペロブスカイト構造 SrRuO_3 は得られない。これは、 SrRuO_3 のみならず、 CaRuO_3 、 BaRuO_3 、その他のペロブスカイト酸化物の場合でも同様である。

【0015】そこで、本発明は、 Si 基板上あるいは Si 基板上のフルオライト構造を有するバッファ層の上に、(100)配向したペロブスカイト構造の酸化物電極薄膜を提供することを目的としている。

【0016】

【課題を解決するための手段】請求項1記載の酸化物電極薄膜は、(100) Si 基板と、前記 Si 基板上に少なくとも NaCl 構造の(100)配向した金属酸化物 MO ($\text{M}=\text{Ca}$ 、 Sr 、 Ba)を含むバッファ層と、前記バッファ層上にペロブスカイト構造の擬立方晶で(100)配向に相当する金属酸化物層 ABO_3 とを有することを特徴とする。

【0017】請求項2記載の酸化物電極薄膜は、フルオライト構造の(100)配向した金属酸化物からなる第1バッファ層と、前記第1バッファ層上に NaCl 構造の(100)配向した金属酸化物 MO ($\text{M}=\text{Ca}$ 、 Sr 、 Ba)からなる第2バッファ層とからなるバッファ層を有することを特徴とする。ただし、フルオライト構造の第1バッファ層は省略し、 NaCl 構造のバッファ層だけにすることも可能である。

【0018】請求項3記載の酸化物電極薄膜は、第1バッファ層において、(100)配向したイットリア安定化ジルコニア $\text{Zr}_{1-x}\text{Y}_x\text{O}_{2-0.5x}$ ($0 < x < 0.45$) と(100)配向した酸化セリウム CeO_2 からなることを特徴とする。

【0019】請求項4記載の酸化物電極薄膜は、金属酸化物層 ABO_3 において、 MRuO_3 ($\text{M}=\text{Ca}$ 、 Sr 、 Ba)からなることを特徴とする。

【0020】請求項5記載の酸化物電極薄膜は、金属酸化物層 ABO_3 において、 $\text{La}_{1-x}\text{Sr}_x\text{VO}_3$ ($0.23 < x < 1$) からなることを特徴とする。

【0021】請求項6記載の酸化物電極薄膜は、金属酸化物層 ABO_3 において、 $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ ($0.2 < x < 0.4$) からなることを特徴とする。

【0022】請求項7記載の酸化物電極薄膜は、金属酸化物層 ABO_3 において、 $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ ($0.4 < x < 0.6$) からなることを特徴とする。

【0023】

【発明の実施の形態】以下、本発明の実施形態を図面に基づいて説明する。

【0024】(実施例1) 図1は本発明の酸化物電極薄膜の第1の実施例を示す図である。

【0025】 YSZ ターゲットを用いたレーザーアブレーション法により、基板温度700℃、酸素分圧 1×1

0^{-6} Torr の条件で、Si 基板 1 上にフルオライト構造の YSZ バッファ層 2 を 50 nm 堆積した。ただし、基板温度、酸素分圧は、これに限るものではない。

【0026】次に、SrO ターゲットを用いたレーザーアブレーション法により、基板温度 700℃、酸素分圧 3mTorr の条件で、NaCl 構造の SrO バッファ層 3 を 10 nm 堆積した。ただし、基板温度、酸素分圧は、これに限るものではない。

【0027】さらに、SrRuO₃ ターゲットを用いたレーザーアブレーション法により、基板温度 700℃、酸素分圧 3mTorr の条件で、ペロブスカイト構造の SrRuO₃ 電極層 4 を 50 nm 堆積した。ただし、基板温度、酸素分圧は、これに限るものではない。

【0028】その後、PbZr_{0.5}Ti_{0.5}O₃ ターゲットを用いたレーザーアブレーション法により、基板温度 700℃、酸素分圧 3mTorr の条件で、ペロブスカイト構造の PbZr_{0.5}Ti_{0.5}O₃ 強誘電体層 5 を 100 nm 堆積した。ただし、基板温度、酸素分圧は、これに限るものではない。

【0029】最後に、SrRuO₃ ターゲットを用いたレーザーアブレーション法により、基板温度 700℃、酸素分圧 3mTorr の条件で、ペロブスカイト構造の SrRuO₃ 上電極層 6 を 50 nm 堆積した。ただし、基板温度、酸素分圧は、これに限るものではない。

【0030】得られた薄膜キャパシタは、膜面に垂直方向で (100) SrRuO₃ / (001) PZT / (100) SrRuO₃ / (100) SrO / (100) YSZ / (100) Si、面内で [110] SrRuO₃ // [110] PZT // [110] SrRuO₃ // [100] SrO // [100] YSZ // [100] Si であった。

【0031】上電極表面での電気抵抗率を 4 端子法によって測定したところ、 $=200 \mu\text{cm}$ の値が得られ、50 K から室温までの広い温度範囲にわたって金属的な温度依存性を示した。

【0032】得られた強誘電体キャパシタについて、周波数 1 kHz、振幅 100 kV/cm の電界を印加してヒステリシス測定を行った結果、 $P_r = 50 \text{ C/cm}^2$ 、 $E_c = 30 \text{ kV/cm}$ の特性が得られ、分極反転 10^{12} 回までファティグによる P_r の劣化は起こらなかった。これは、(100) Si 基板上に (100) YSZ バッファ層を介してエピタキシャル成長させた (110) PZT 薄膜を有する強誘電体キャパシタの場合の $P_r = 30 \text{ C/cm}^2$ 、 $E_c = 50 \text{ kV/cm}$ 、ファティグ特性 10^{11} 回と比較して、強誘電体メモリとしてのより高い電気特性を有している。

【0033】上述のように、フルオライト構造の YSZ および NaCl 構造の SrO からなる二重バッファ層を用いて Si 基板上に (100) 配向ペロブスカイト酸化物電極薄膜を堆積することにより、強誘電体キャパシタ

の P_r 、 E_c 、ファティグ特性を向上させることが可能となる。

【0034】(実施例 2) 図 2 は本発明の酸化物電極薄膜の第 2 の実施例を示す図である。

【0035】SrO ターゲットを用いたレーザーアブレーション法により、基板温度 700℃、酸素分圧 1×10^{-6} Torr の条件で、Si 基板上 21 上に NaCl 構造の SrO バッファ層 22 を 10 nm 堆積した。ただし、基板温度、酸素分圧は、これに限るものではない。

【0036】さらに、La_{0.7}Sr_{0.3}MnO₃ ターゲットを用いたレーザーアブレーション法により、基板温度 700℃、酸素分圧 3mTorr の条件で、ペロブスカイト構造の La_{0.7}Sr_{0.3}MnO₃ 下電極層 23 を 50 nm 堆積した。ただし、基板温度、酸素分圧は、これに限るものではない。

【0037】その後、PbZr_{0.5}Ti_{0.5}O₃ ターゲットを用いたレーザーアブレーション法により、基板温度 700℃、酸素分圧 3mTorr の条件で、ペロブスカイト構造の PbZr_{0.5}Ti_{0.5}O₃ 強誘電体層 24 を 100 nm 堆積した。ただし、基板温度、酸素分圧は、これに限るものではない。

【0038】最後に、La_{0.7}Sr_{0.3}MnO₃ ターゲットを用いたレーザーアブレーション法により、基板温度 700℃、酸素分圧 3mTorr の条件で、ペロブスカイト構造の La_{0.7}Sr_{0.3}MnO₃ 上電極層 25 を 50 nm 堆積した。ただし、基板温度、酸素分圧は、これに限るものではない。

【0039】得られた薄膜キャパシタは、膜面に垂直方向で (100) La_{0.7}Sr_{0.3}MnO₃ / (001) PZT / (100) La_{0.7}Sr_{0.3}MnO₃ / (100) SrO / (100) YSZ / (100) Si、面内で [110] La_{1-x}Sr_xMnO₃ // [110] PZT // [110] La_{1-x}Sr_xMnO₃ // [100] SrO // [100] YSZ // [100] Si であった。

【0040】ここで La_{1-x}Sr_xMnO₃ において $x = 0.3$ としたが、 0.2×0.4 であれば、同様の効果が得られる。

【0041】得られた強誘電体キャパシタについて、周波数 1 kHz、振幅 100 kV/cm の電界を印加してヒステリシス測定を行った結果、 $P_r = 45 \text{ C/cm}^2$ 、 $E_c = 30 \text{ kV/cm}$ の高い電気特性が得られ、分極反転 10^{12} 回までファティグによる P_r の劣化は起こらなかった。これは、(100) Si 基板上に直接堆積させた PZT 多結晶薄膜を有する強誘電体キャパシタの場合の $P_r = 10 \text{ C/cm}^2$ 、 $E_c = 100 \text{ kV/cm}$ 、ファティグ特性 10^8 回と比較して、強誘電体メモリとしてのより高い電気特性を有している。

【0042】また、La_{1-x}Sr_xMnO₃ の代わりに La_{1-x}Sr_xVO₃ (0.23×1)、La_{1-x}Sr_xCo

O_3 (0.4 x 0.6) を電極薄膜に用いた場合も同様の効果が得られる。

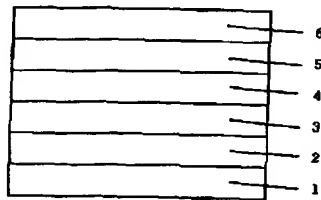
【0043】 上述のように、NaCl 構造の SrO バッファ層を用いて Si 基板上に (100) 配向ペロブスカイト酸化物電極薄膜を堆積することにより、強誘電体キャパシタの P_r 、 E_c 、ファティーグ特性を向上させることが可能となる。

【0044】

【発明の効果】 以上述べたように、本発明の酸化物電極薄膜によれば、シリコン基板上に、少なくとも最上面に NaCl 構造を有するバッファ層を形成することにより、そのバッファ層上にペロブスカイト構造の酸化物電極薄膜を (100) あるいは (001) 配向で成長させることができ、誘電体キャパシタメモリ、強誘電体キャパシタメモリ、MFSFET などの電子デバイスを最適な構造で実現することができる。

【図面の簡単な説明】

【図 1】



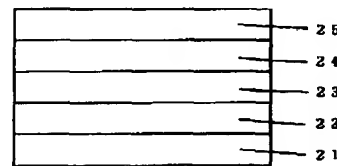
【図 1】 本発明の一実施例を示す SrRuO_3 酸化物電極薄膜を含んだ強誘電体キャパシタの断面図である。

【図 2】 本発明の一実施例を示す $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ 酸化物電極薄膜を含んだ強誘電体キャパシタの断面図である。

【符号の説明】

1. Si 基板
2. YSZ バッファ層
3. SrO バッファ層
4. SrRuO_3 下電極層
5. PZT 強誘電体層
6. SrRuO_3 上電極層
21. Si 基板
22. SrO バッファ層
23. $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ 下電極層
24. PZT 強誘電体層
25. $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ 上電極層

【図 2】



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 CLAIMS

[Claim(s)]

[Claim 1] (100) The oxide electrode thin film characterized by having a silicon substrate, a buffer layer containing the metallic oxide MO (M=calcium, Sr, Ba) in which NaCl structure carried out orientation (100) at least on the aforementioned silicon substrate, and the metallic-oxide layer ABO₃ that is equivalent to orientation (100) by the pseudo-cubic of a perovskite structure on the aforementioned buffer layer.

[Claim 2] The aforementioned buffer layer is an oxide electrode thin film according to claim 1 characterized by the bird clapper from the 1st buffer layer which consists of a metallic oxide in which fluorite structure carried out orientation (100), and the 2nd buffer layer which consists of a metallic oxide MO (M=calcium, Sr, Ba) in which NaCl structure carried out orientation (100) on the 1st buffer layer of the above.

[Claim 3] The 1st buffer layer of the above is an oxide electrode thin film according to claim 2 characterized by the bird clapper from the cerium oxide CeO₂ which carried out orientation (100) to yttria-stabilized-zirconia Zr_{1-x}Y_xO_{2-0.5x} (0<x<0.45) which carried out orientation (100).

[Claim 4] The aforementioned metallic-oxide layer ABO₃ is an oxide electrode thin film according to claim 1 characterized by the bird clapper from MRuO₃ (M=calcium, Sr, Ba).

[Claim 5] The aforementioned account metallic-oxide layer ABO₃ is an oxide electrode thin film according to claim 1 characterized by the bird clapper from La_{1-x}Sr_xVO₃ (0.23<x<1).

[Claim 6] The aforementioned metallic-oxide layer ABO₃ is an oxide electrode thin film according to claim 1 characterized by the bird clapper from La_{1-x}Sr_xMnO₃ (0.2<x<0.4).

[Claim 7] The aforementioned account metallic-oxide layer ABO₃ is an oxide electrode thin film according to claim 1 characterized by the bird clapper from La_{1-x}Sr_xCoO₃ (0.4<x<0.6).

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] Especially this invention relates to oxide devices, such as ferroelectric random-access memory using this oxide electrode thin film, about an oxide electrode thin film.

[0002]

[Description of the Prior Art] In recent years, development of the non-volatile memory using the ferroelectric is progressing quickly. As a ferroelectric, although material, such as $\text{PbZr}_{1-x}\text{Ti}_x\text{O}_3$ (PZT), $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ (BST), and $\text{SrBi}_2\text{Ta}_2\text{O}_9$ (SBT), attracts attention, it is the material to which the PZT material of composition near a rhombohedron-tetragonal phase boundary (MPB) is excellent in the remanence and the anti-electric-field property, and utilization is progressing most especially.

[0003] Ferroelectric random-access memory is classified into the ferroelectric capacitor type which forms 1T / 1C structure, using a ferroelectric as a capacitor, and the MFSFET type which uses a ferroelectric as a gate insulator layer instead of SiO_2 . In the ferroelectric capacitor structure of 1T/1C type, since an electrode layer will be formed on substrates, such as silicon (Si), and a ferroelectric layer will be formed on it, selection of an electrode material has big influence on the property of a ferroelectric. For the property demanded as an electrode material of a ferroelectric capacitor, (1) electric resistance is a low enough.

(2) The mismatch of a lattice constant with ferroelectric material is small.

(3) Thermal resistance is high.

(4) Reactivity is a low.

(5) Diffusion barrier property is high.

(6) Adhesion with a substrate and a ferroelectric is good.

It is ****.

[0004] Conventionally, Pt has been used as an electrode material of a PZT system ferroelectric. If a self-stacking tendency is strong and forms membranes on [like SiO_2] amorphous in order to take the face-centered cubic lattice (FCC) structure which is a close packed structure (111), orientation of the Pt will be carried out strongly, and the ferroelectric film on it also has a good stacking tendency. However, since the stacking tendency was strong, the columnar crystal grew, and there was a problem of becoming easy to diffuse Pb etc. on a ground along with a grain boundary. Moreover, there was a problem also in the adhesion of Pt and SiO_2 .

[0005] For an improvement of the adhesion of Pt and SiO_2 , Ti and when [even if it uses TiN etc. as diffusion barrier layers, such as Pb further,] becoming complicated electrode structure, oxidization of Ti, diffusion into Pt of Ti, and the crystalline fall of PZT accompanying it take place here, and electrical properties, such as an P-E hysteresis characteristic, a leakage-current property, and a fatigue property, deteriorate.

[0006] Thus, since there are many problems in Pt electrode, RuO_x and IrO_2 are begun and the conductive oxide electrode material is studied. Especially, since it has the same crystal structure as PZT,

SrRuO₃ which has a perovskite structure is excellent in the junction nature in an interface, tends to realize epitaxial growth of PZT and is excellent also in the property as a diffusion barrier layer of Pb. Therefore, research of the ferroelectric capacitor using SrRuO₃ as an electrode is done briskly.

[0007]

[Problem(s) to be Solved by the Invention] However, when a ferroelectric capacitor was built by using PZT as a ferroelectric, having used SrRuO₃ electrode as the oxide electrode thin film, there were the following problems.

[0008] Composition of the overTi side, Zr:Ti=0.3:0.7 [for example,], and 0.2:0.8 is more important for PZT than MPB of Zr:Ti=0.48:0.52 from increase of Remanence Pr, and the point of reduction of the anti-electric field Ec. Moreover, PZT in this composition field shows a tetragonal phase, and the direction of polarization is parallel to c axis. Therefore, in order to obtain the orientation (001) film of the ferroelectric layer PZT, it is necessary to make a direction (100) (pseudo-cubic) carry out orientation growth of the SrRuO₃ electrode itself which is a lower electrode in the ferroelectric capacitor which has the structure of an up electrode / ferroelectric / lower electrode / substrate.

[0009] However, it is difficult to grow epitaxially directly SrRuO₃ electrode which is a perovskite type oxide on Si. Then, it is necessary to grow a certain buffer layer epitaxially on Si, and to grow SrRuO₃ electrode epitaxially on it.

[0010] Here, as a buffer layer for growing epitaxially YBa₂Cu₃O_x which has perovskite and analogous structure, the oxide which has fluorite structures, such as yttria-stabilized-zirconia Zr_{1-x}Y_xO_{2-0.5x} (YSZ), CeO₂, and Y₂O₃, has been used on Si. Moreover, it is confirmed that Y₂O₃/the double-buffers layer of CeO₂ [YSZ ((1) Appl.Phys.Lett. and 61 1240 (1992)) and]/YSZ ((2) Appl.Phys.Lett. and 64 1573 (1994)) also grows YBa₂Cu₃O_x epitaxially. In this case, orientation of the YBa₂Cu₃O_x is carried out easily (001).

[0011] However, in SrRuO₃ (it sets to a pseudo-cubic and is $a = 0.556\text{nm}$ $2^{1/2}$ $a = 0.393\text{nm}$) which is simple perovskite structure, on YSZ ($a = 0.514\text{nm}$) of fluorite type structure, or the field (100) of CeO₂ ($a = 0.541\text{nm}$), although the mismatching of a grid is as small as about 3% at the case of CeO₂, it does not grow epitaxially in a direction (100). this is a field where the charge of only oxygen ion is not neutralized [Ce ion] for the maximum front face in the fluorite structure of orientation (100) -- receiving -- the perovskite-type structure of orientation (100) -- every -- it is the field where the charge which AO and BO two-layer become from a cation and an anion was neutralized, and is for repulsive force to arise by the electrostatic interaction of the maximum front face of a ground, and the first pass of a deposit

[0012] For example, on the YSZ (100) substrate, carrying out orientation (pseudo-cubic) (110) is known ((3) Appl.Phys.Lett. and 67 1387-1389 (1995)). When the perovskite structure of orientation (110) carries out a laminating to the maximum front face of the fluorite structure of orientation (100), this is structure in which an ABO side and O₂ page carry out a laminating in the direction of a field (110), and the charge of each side does not neutralize but it is considered to carry out orientation easily on the fluorite structure of orientation (100) in the viewpoint of an electrostatic interaction, since it is stable.

[0013] Furthermore, the oxide laminated structure and the manufacture method of SrRuO₃ (100)₃ / (100) CeO₂/(100) Si are proposed ((4) JP,10-182292,A). However, in fact, epitaxial growth of CeO₂ (100)₂/ (100) Si is possible only under the elevated temperature of 600 degrees C or more, and the high-vacuum atmosphere of 10 to 8 or less Torrs, and moreover does not become a single orientation film, but coexists with the field of CeO₂ (110)₂/(100) Si ((5) Jpn.J.Appl.Phys., 29, and L1199 (1990)). Moreover (100), the direction (110) and the direction (100) live together also in SrRuO₃ of SrRuO₃/(100) CeO₂/ (100) YSZ/(100)Si.

[0014] As mentioned above, as long as simultaneous supply of Sr and the Ru is carried out and they grow, the perovskite structure SrRuO₃ of orientation (100) is not obtained on Si substrate or the buffer layer of fluorite structure. This is the same not only at SrRuO₃ but the case of CaRuO₃, BaRuO₃, and other perovskite oxides.

[0015] Then, this invention aims at offering the oxide electrode thin film of the perovskite structure which carried out orientation (100) on the buffer layer which has the fluorite structure on Si substrate or

Si substrate.

[0016]

[Means for Solving the Problem] An oxide electrode thin film according to claim 1 is characterized by having Si (100) substrate, a buffer layer containing the metallic oxide MO (M=calcium, Sr, Ba) in which NaCl structure carried out orientation (100) at least on the aforementioned Si substrate, and the metallic-oxide layer ABO₃ that is equivalent to orientation (100) by the pseudo-cubic of a perovskite structure on the aforementioned buffer layer.

[0017] An oxide electrode thin film according to claim 2 is characterized by having the buffer layer which consists of the 1st buffer layer which consists of a metallic oxide in which fluorite structure carried out orientation (100), and the 2nd buffer layer which consists of a metallic oxide MO (M=calcium, Sr, Ba) in which NaCl structure carried out orientation (100) on the 1st buffer layer of the above. However, it is also possible for the 1st buffer layer of fluorite structure to omit and to make it only the buffer layer of NaCl structure.

[0018] An oxide electrode thin film according to claim 3 is characterized by the bird clapper in the 1st buffer layer from the cerium oxide CeO₂ which carried out orientation (100) to yttria-stabilized-zirconia Zr_{1-x}Y_xO_{2-0.5x} (0<x<0.45) which carried out orientation (100).

[0019] An oxide electrode thin film according to claim 4 is characterized by the bird clapper from MRuO₃ (M=calcium, Sr, Ba) in the metallic-oxide layer ABO₃.

[0020] An oxide electrode thin film according to claim 5 is characterized by the bird clapper from La_{1-x}Sr_xVO₃ (0.23<x<1) in the metallic-oxide layer ABO₃.

[0021] An oxide electrode thin film according to claim 6 is characterized by the bird clapper from La_{1-x}Sr_xMnO₃ (0.2<x<0.4) in the metallic-oxide layer ABO₃.

[0022] An oxide electrode thin film according to claim 7 is characterized by the bird clapper from La_{1-x}Sr_xCoO₃ (0.4<x<0.6) in the metallic-oxide layer ABO₃.

[0023]

[Embodiments of the Invention] Hereafter, the operation gestalt of this invention is explained based on a drawing.

[0024] (Example 1) Drawing 1 is drawing showing the 1st example of the oxide electrode thin film of this invention.

[0025] By the laser ablation method using the YSZ target, 50nm of YSZ buffer layers 2 of fluorite structure was deposited on the Si substrate 1 on condition that substrate temperature [of 700 degrees C], and oxygen tension 1x10⁻⁶Torr. However, substrate temperature and oxygen tension are not restricted to this.

[0026] Next, 10nm of SrO buffer layers 3 of NaCl structure was deposited by the laser ablation method using the SrO target on condition that the substrate temperature of 700 degrees C, and oxygen tension 3mTorr. However, substrate temperature and oxygen tension are not restricted to this.

[0027] Furthermore, 50nm of SrRuO₃ electrode layers 4 of a perovskite structure was deposited by the laser ablation method using SrRuO₃ target on condition that the substrate temperature of 700 degrees C, and oxygen tension 3mTorr. However, substrate temperature and oxygen tension are not restricted to this.

[0028] Then, 100nm of 0.5O₃ or so PbZr_{0.5}Ti dielectric layers 5 of a perovskite structure was deposited by the laser ablation method using 0.5OPbZr_{0.5}Ti₃ target on condition that the substrate temperature of 700 degrees C, and oxygen tension 3mTorr. However, substrate temperature and oxygen tension are not restricted to this.

[0029] Finally, 50nm of SrRuO₃ top electrode layers 6 of a perovskite structure was deposited by the laser ablation method for having used SrRuO₃ target on condition that the substrate temperature of 700 degrees C, and oxygen tension 3mTorr. However, substrate temperature and oxygen tension are not restricted to this.

[0030] The obtained thin film capacitor was perpendicular to the film surface, and were SrO//[100] YSZ//[[110] PZT / [110] SrRuO₃////[110] SrRuO₃// [100]] [100]Si in SrRuO₃(100)₃/(001) PZT/(100) SrRuO₃/(100) SrO/(100) YSZ/(100)Si, and the field.

[0031] When the electrical resistivity in an upper electrode front face was measured by 4 terminal method, the value of $\approx 200 \mu\Omega\text{cm}$ was acquired and the temperature dependence like a metal was shown over the latus temperature requirement from 50K to a room temperature.

[0032] As a result of impressing the electric field of the frequency of 1kHz, and amplitude 100 kV/cm and performing hysteresis measurement about the obtained ferroelectric capacitor, the property ($P_r=50 \text{ C/cm}^2$ and $E_c=30 \text{ kV/cm}$) was acquired, and degradation of P_r by the fatigue did not take place to 1012 polarization reversal. This has the higher electrical property as ferroelectric random-access memory as compared with $P_r=30 \text{ C/cm}^2$ in the case of the ferroelectric capacitor which has the PZT (110) thin film grown epitaxially through the YSZ (100) buffer layer on Si (100) substrate, $E_c=50 \text{ kV/cm}$, and 1011 fatigue properties.

[0033] As mentioned above, it becomes possible to raise P_r of a ferroelectric capacitor, E_c , and a fatigue property by depositing an orientation (100) perovskite oxide electrode thin film on Si substrate using the double-buffers layer which consists of YSZ of fluorite structure, and SrO of NaCl structure.

[0034] (Example 2) Drawing 2 is drawing showing the 2nd example of the oxide electrode thin film of this invention.

[0035] By the laser ablation method using the SrO target, 10nm of SrO buffer layers 22 of NaCl structure was deposited on Si substrate top 21 on condition that substrate temperature [of 700 degrees C], and oxygen tension 1×10^{-6} Torr. However, substrate temperature and oxygen tension are not restricted to this.

[0036] Furthermore, 50nm of bottom electrode layers 23 of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ of a perovskite structure was deposited by the laser ablation method using $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ target on condition that the substrate temperature of 700 degrees C, and oxygen tension 3mTorr. However, substrate temperature and oxygen tension are not restricted to this.

[0037] Then, 100nm of 0.5O_3 or so $\text{PbZr}_{0.5}\text{Ti}$ dielectric layers 24 of a perovskite structure was deposited by the laser ablation method using $0.5\text{OPbZr}_{0.5}\text{Ti}_3$ target on condition that the substrate temperature of 700 degrees C, and oxygen tension 3mTorr. However, substrate temperature and oxygen tension are not restricted to this.

[0038] Finally, 50nm of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ top electrode layers 25 of a perovskite structure was deposited by the laser ablation method for having used $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ target on condition that the substrate temperature of 700 degrees C, and oxygen tension 3mTorr. However, substrate temperature and oxygen tension are not restricted to this.

[0039] The obtained thin film capacitor is perpendicular to a film surface. $\text{La}(100)_{0.7}\text{Sr}_{0.3}\text{MnO}_3/(001) \text{PZT}/(100) \text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3/(100) \text{SrO}/(100) \text{YSZ}/(100) \text{Si}$, They were $\text{SrO}//[100] \text{YSZ}//[110] \text{PZT} / [110] \text{La}_{1-x}\text{Sr}_x\text{MnO}_3////[110] \text{La}_{1-x}\text{Sr}_x\text{MnO}_3/[100] [100] \text{Si}$ in the field.

[0040] Although referred to as $x=0.3$ in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ here, the same effect will be acquired if it is 0.2×0.4 .

[0041] As a result of impressing electric field with a frequency [of 1kHz], and an amplitude of 100kV [/cm] and performing hysteresis measurement about the obtained ferroelectric capacitor, the high electrical property of $P_r=45 \text{ C/cm}^2$ and $E_c=30 \text{ kV/cm}$ was acquired, and degradation of P_r by the fatigue did not take place to 1012 polarization reversal. This has the higher electrical property as ferroelectric random-access memory as compared with $P_r=10 \text{ C/cm}^2$ in the case of the ferroelectric capacitor which has the PZT polycrystal thin film made to deposit directly on Si (100) substrate, $E_c=100 \text{ kV/cm}$, and 108 fatigue properties.

[0042] Moreover, the same effect is acquired when $\text{La}_{1-x}\text{Sr}_x\text{VO}_3$ (0.23×1) and $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ (0.4×0.6) are used for an electrode thin film instead of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$.

[0043] As mentioned above, it becomes possible by depositing an orientation (100) perovskite oxide electrode thin film on Si substrate using the SrO buffer layer of NaCl structure to raise P_r of a ferroelectric capacitor, E_c , and a fatigue property.

[0044]

[Effect of the Invention] forming at least the buffer layer which was described above and which has NaCl structure in the best side on a silicon substrate like according to the oxide electrode thin film of

this invention -- the buffer-layer top -- the oxide electrode thin film of a perovskite structure -- or (100) (001) it can be made to be able to grow up by orientation and electron devices, such as dielectric capacitor memory, ferroelectric capacitor memory, and MFSFET, can be realized with the optimal structure

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PRIOR ART

[Description of the Prior Art] In recent years, development of the non-volatile memory using the ferroelectric is progressing quickly. As a ferroelectric, although material, such as $\text{PbZr}_{1-x}\text{Ti}_x\text{O}_3$ (PZT), $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ (BST), and $\text{SrBi}_2\text{Ta}_2\text{O}_9$ (SBT), attracts attention, it is the material to which the PZT material of composition near a rhombohedron-tetragonal phase boundary (MPB) is excellent in the remanence and the anti-electric-field property, and utilization is progressing most especially.

[0003] Ferroelectric random-access memory is classified into the ferroelectric capacitor type which forms 1T / 1C structure, using a ferroelectric as a capacitor, and the MFSFET type which uses a ferroelectric as a gate insulator layer instead of SiO_2 . In the ferroelectric capacitor structure of 1T/1C type, since an electrode layer will be formed on substrates, such as silicon (Si), and a ferroelectric layer will be formed on it, selection of an electrode material has big influence on the property of a ferroelectric. For the property demanded as an electrode material of a ferroelectric capacitor, (1) electric resistance is a low enough.

(2) The mismatch of a lattice constant with ferroelectric material is small.

(3) Thermal resistance is high.

(4) Reactivity is a low.

(5) Diffusion barrier property is high.

(6) Adhesion with a substrate and a ferroelectric is good.

It is ****.

[0004] Conventionally, Pt has been used as an electrode material of a PZT system ferroelectric. If a self-stacking tendency is strong and forms membranes on [like SiO_2] amorphous in order to take the face-centered cubic lattice (FCC) structure which is a close packed structure (111), orientation of the Pt will be carried out strongly, and the ferroelectric film on it also has a good stacking tendency. However, since the stacking tendency was strong, the columnar crystal grew, and there was a problem of becoming easy to diffuse Pb etc. on a ground along with a grain boundary. Moreover, there was a problem also in the adhesion of Pt and SiO_2 .

[0005] For an improvement of the adhesion of Pt and SiO_2 , Ti and when [even if it uses TiN etc. as diffusion barrier layers, such as Pb further,] becoming complicated electrode structure, oxidization of Ti, diffusion into Pt of Ti, and the crystalline fall of PZT accompanying it take place here, and electrical properties, such as an P-E hysteresis characteristic, a leakage-current property, and a fatigue property, deteriorate.

[0006] Thus, since there are many problems in Pt electrode, RuO_x and IrO_2 are begun and the conductive oxide electrode material is studied. Especially, since it has the same crystal structure as PZT, SrRuO_3 which has a perovskite structure is excellent in the junction nature in an interface, tends to realize epitaxial growth of PZT and is excellent also in the property as a diffusion barrier layer of Pb. Therefore, research of the ferroelectric capacitor using SrRuO_3 as an electrode is done briskly.

[Translation done.]

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] It is the cross section of the ferroelectric capacitor containing the SrRuO teroxide electrode thin film which shows one example of this invention.

[Drawing 2] It is the cross section of the ferroelectric capacitor containing the La_{0.7}Sr_{0.3}MnO teroxide electrode thin film which shows one example of this invention.

[Description of Notations]

1. Si Substrate
2. YSZ Buffer Layer
3. SrO Buffer Layer
4. Bottom Electrode Layer of SrRuO₃
5. PZT Ferroelectric Layer
6. SrRuO₃ Top Electrode Layer
21. Si Substrate
22. SrO Buffer Layer
23. Bottom Electrode Layer of La_{0.7}Sr_{0.3}MnO₃
24. PZT Ferroelectric Layer
25. La_{0.7}Sr_{0.3}MnO₃ Top Electrode Layer

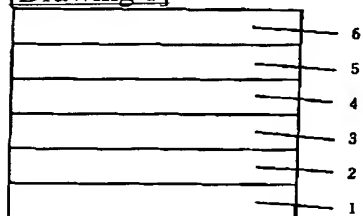
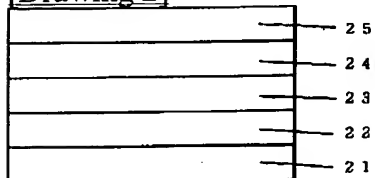
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DRAWINGS

[Drawing 1][Drawing 2]

[Translation done.]